
Speciation of Cs-137 and Pu Isotopes in Polessie State Radiation-Ecological Reserve Soil, Chernobyl Zone.

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Abstract Even though a huge effort has been expended on determining levels of radioactive contamination following the Chernobyl nuclear power plant accident in 1986, within the Belarusian sector of the Chernobyl exclusion zone, information has been inadequate. Speciation, transfer and fate of numerous radionuclides most notably transuranium radioisotopes within in this region are poorly understood. In this study, the main goal was to investigate if forest fires could change the speciation of the radionuclides present in the tops soil within the Polessie State Radiation-Ecological Reserve (PSRER), within Belarusian side of the Chernobyl exclusion zone. Even though no changes in speciation can be seen for Cs, a slight change for the Pu-isotopes can be seen comparing soils from inside and outside forest fire areas. Within the forest fire area, the Pu-isotopes are more bound to the more oxidiseable form, and if incorporated in particles, this will be more susceptible to weathering after a fire.

1. INTRODUCTION

Even though a huge effort has been expended on determining levels of radioactive contamination following the Chernobyl nuclear power plant accident in 1986, within the Belarusian sector of the Chernobyl exclusion zone, information has been inadequate. Speciation, transfer and fate of numerous radionuclides most notably transuranium radioisotopes within this region are poorly understood. After deposition, weathering of radioactive particles and mobilization of associated radionuclides occurs with time. The transfer of mobilized radionuclides within the ecosystem will be delayed and depends on the weathering rate. The weathering rate in turn depends upon the initial particle composition, structural changes occurring during the event e.g., transformation from UO_2 to U_3O_8 and U_2O_5 , and transformation processes taking place after deposition depending on soil pH, redox conditions and microbial activity [1].

The Chernobyl accident resulted in the release and dispersion of about 3 % of the irradiated uranium dioxide fuel in the form of “hot particles” of various sizes and degree of oxidation. The ^{90}Sr , that normally is quite mobile, showed a low migration mobility when it was incorporated into fuel particles. However, as fuel particles dissolve due to weathering, the portion of extractable ^{90}Sr in soils increase, leading to higher soil-to-vegetation transfer coefficients. The released fuel particles can be categorized roughly into “non-oxidized” (from the explosion) and “oxidized” (from the following fire) types. Of the total release from Chernobyl NPP, more than 90 % of the radiologically important

radionuclides (^{90}Sr , Pu-isotopes, ^{241}Am) were in the form of small (less than 10 μm diameter) fuel particles. Within the 30-km zone, a major fraction of the deposited radionuclides, including ^{137}Cs , were incorporated within fuel particle matrices, hence contamination levels of ^{90}Sr and transuranics were relatively high [2]. In the case of forest fires, the speciation of the deposited fuel particles can be changed due to oxidizing conditions. This might lead to increase in weathering rates in forest fire areas.

The association of radionuclides with soil components will influence their subsequent transfer to compartments in the environment, and thereby their mobility in soils and availability for biological uptake by vegetation. In this study, soil from within and outside of areas affected by forest fire have been subjected to sequential extractions in order to identify the association of radionuclides with different soil components and reveal any differences in speciation between the two areas. Sequential extraction can also be used to indicate the presence of fuel particles in soils, due to the leaching of refractory particles with the H_2O_2 fraction.

How the speciation of ^{137}Cs and Pu-isotopes are affected by extreme environmental phenomena such as forest fires is unknown. Forest fires may either change the soil characteristics or act as an oxidizing event in itself. Based on this, the following hypothesis was formulated:

Speciation of ^{137}Cs and Plutonium-isotopes (Pu-isotopes) within and outside of areas affected by forest fires is different, and forest fires are change the particles, leaving them more oxidized and more susceptible to weathering.

2. MATERIAL AND METHODS

Fieldwork was conducted by a joint team from the Norwegian University of Life Science (UMB), Norwegian Radiation Protection Authority (NRPA), Belarus State University and staff from the Polessie State Radiological Ecological Reserve (PSRER). Fieldwork was performed in May 2009 and May 2010 in the PSRER in Gomel, Belarus. Sites of investigation are shown in figure 1.

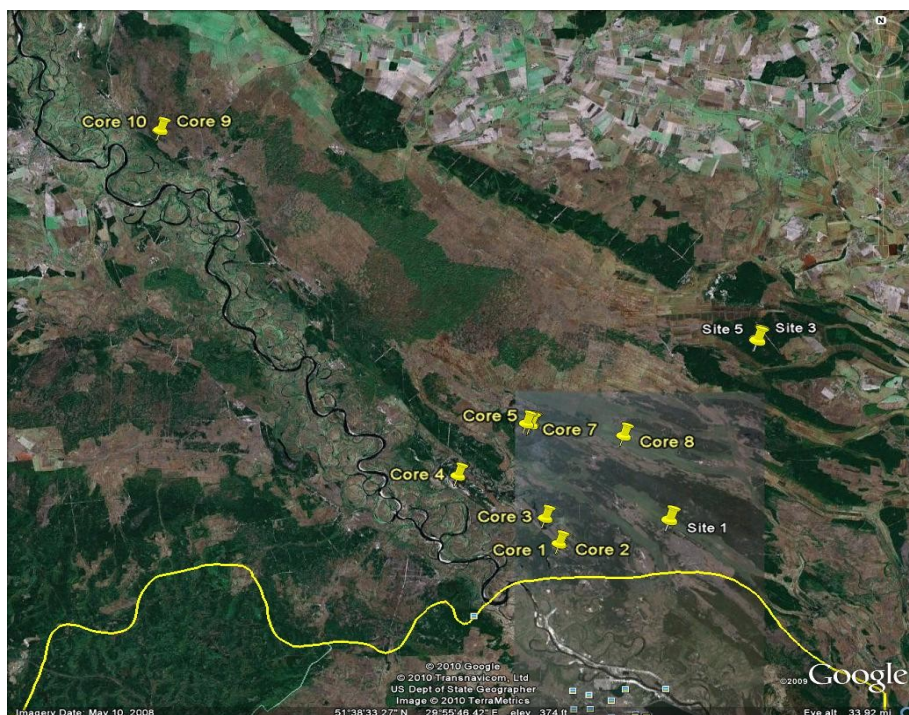


Figure 1. Map showing the different soil samplings sites. Sampling sites within forest fire areas are sites 9 and 10. The other sites are outside fire-affected areas. The yellow line is the border between Belarus and Ukraine.

Soil samples (cores) were taken with a cylindrical borer and divided into 7 layers based on depth below the humus layer; 0-2 cm, 2-4 cm, 4-6 cm, 6-8 cm, 8-10 cm, 10-15 cm and 15-20 cm. After soil characterization (pH, dry matter, LoI, clay content), Cs-137 activity concentrations were determined with a NaI detector. Layers with the highest Cs content were subjected to particle search using repeated splitting of the sample and digital autoradiography (Typhoon 8600, Molecular Dynamics). Pu analysis was performed after radiochemical separation[3] and measured with an alpha spectrometer (model 7401, Canberra Industries). Sequential extraction procedures were performed according to a modified method after Tessier 1979[4; 5].

3. RESULTS AND DISCUSSION

The highest activity concentrations of ^{137}Cs were found in the 0-2 and 2-4 cm depth layers of soil (Figure 2). Activity concentrations in the 0-2 and 2-4 cm layers of all cores (1-10) fell in the range from 2 - 120 Bq/g dry weight, cores outside forest fire affected areas (5-8) were in the range 3 - 120 Bq/g dry weight. The forest fire affected cores (9 and 10) ranged from 7 - 22 Bq/g dry weight. The total ^{137}Cs activity concentrations were in the range of earlier reported values[6].

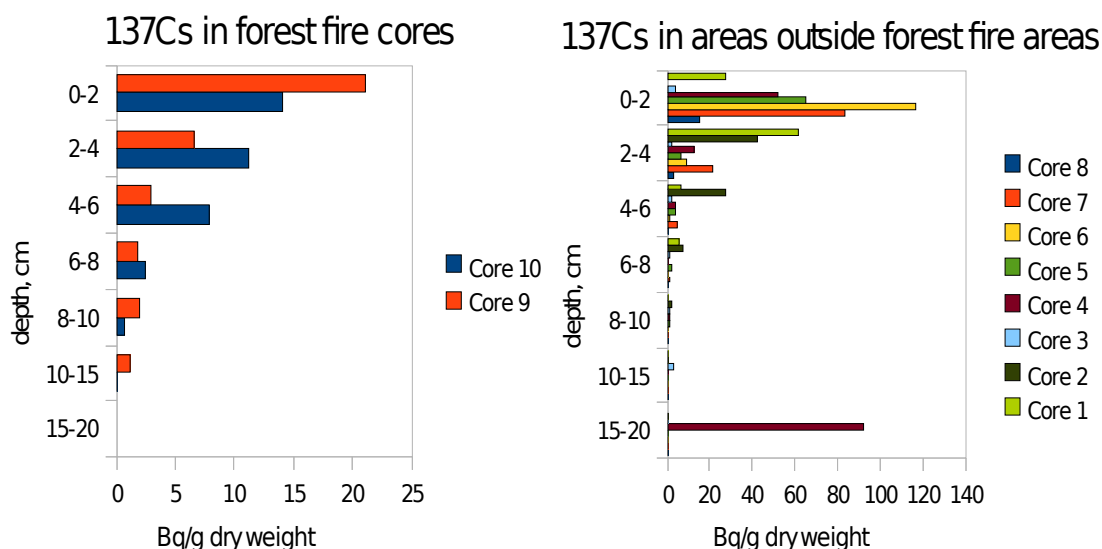


Figure 2. Cs-137 activity concentrations in soil within (left) and outside (right) forest fire affected areas.

Sequential extraction results for ^{137}Cs are shown in figure 3. There was no significant differences in the speciation for soil outside or within forest fire affected areas. For all soil samples, the ^{137}Cs was found mainly in the irreversibly bound (strong acid leachable) fraction (40-65%) or in the residual fraction (35-60%). Overall, the results are consistent with previously published data on radionuclide, supporting a strong association of radionuclides with mineral components in soils[5].

Total Pu activity concentrations are shown in Table 1, The Pu-isotope concentrations in soil outside forest fire affected areas ranged from 6.8 ± 0.4 to 96 ± 3 Bg/kg and 3.6 ± 0.1 to 200 ± 3 Bg/kg, for Pu-238 and Pu-239,240, respectively. Within the forest fire affected areas the activity concentrations for Pu-238 and Pu-239,240, ranged from 13.7 ± 0.6 to 17.9 ± 0.7 and 20.5 ± 0.5 to 27 ± 0.6 Bq/kg, respectively. This was in the same range as measured recently by Ovsianikova et al. [7] .

The speciation of Pu-isotopes for soils from outside and within forest fire affected areas is shown in figure 4. For all soils, Pu isotopes are mostly found to be strongly or irreversibly bound. For soils outside the forest fire affected area, about 20 % is found associated with the irreversibly bound, HNO_3 -extractable fraction while 80% is in the more oxidiseable form (H_2O_2), indicating the presence of particles or association with organic matter. In soils from forest fire affected areas all Pu-isotopes were found to be associated with the H_2O_2 fraction, indicating that fire may have changed the speciation of Pu slightly.

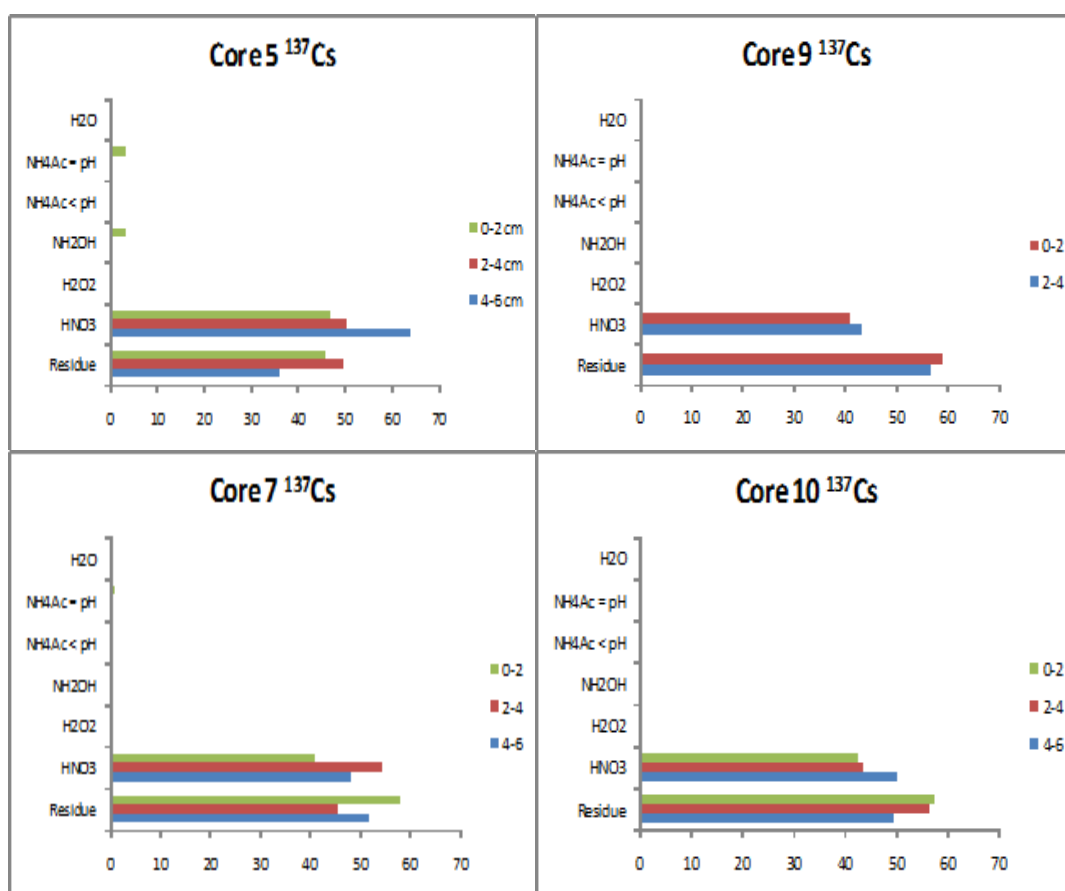


Figure 3 Sequential extraction results of soil outside forest fire area (left, cores 5 and 7) and of soil within fire areas (right, cores 9 and 10).

Table 1. Total Pu-isotope activity concentrations in soils from inside and outside forest fire areas.

Site	Pu-238 Bq/kg (d.w.)	Pu-239,240 Bq/kg (d.w.)
<i>Outside forest fire area</i>		
1	40 ± 2	76 ± 2
4	6.8 ± 0.4	3.6 ± 0.1
6	96 ± 3	200 ± 3
<i>Inside forest fire area</i>		
9	17.9 ± 0.7	27 ± 0.6
10	13.7 ± 0.7	20.5 ± 0.5

The search for hot particles in the areas revealed that, even though high activity levels were detected, the particles in the soil within the forest fire affected area were too small to be isolated, while there were larger particles found outside the forest fire affected areas. This is shown in the digital autoradiography image

reproduced in figure 5. The reason for this difference in particle size, might be due to the forest fire, increasing the weathering rate of the particles in the soil.

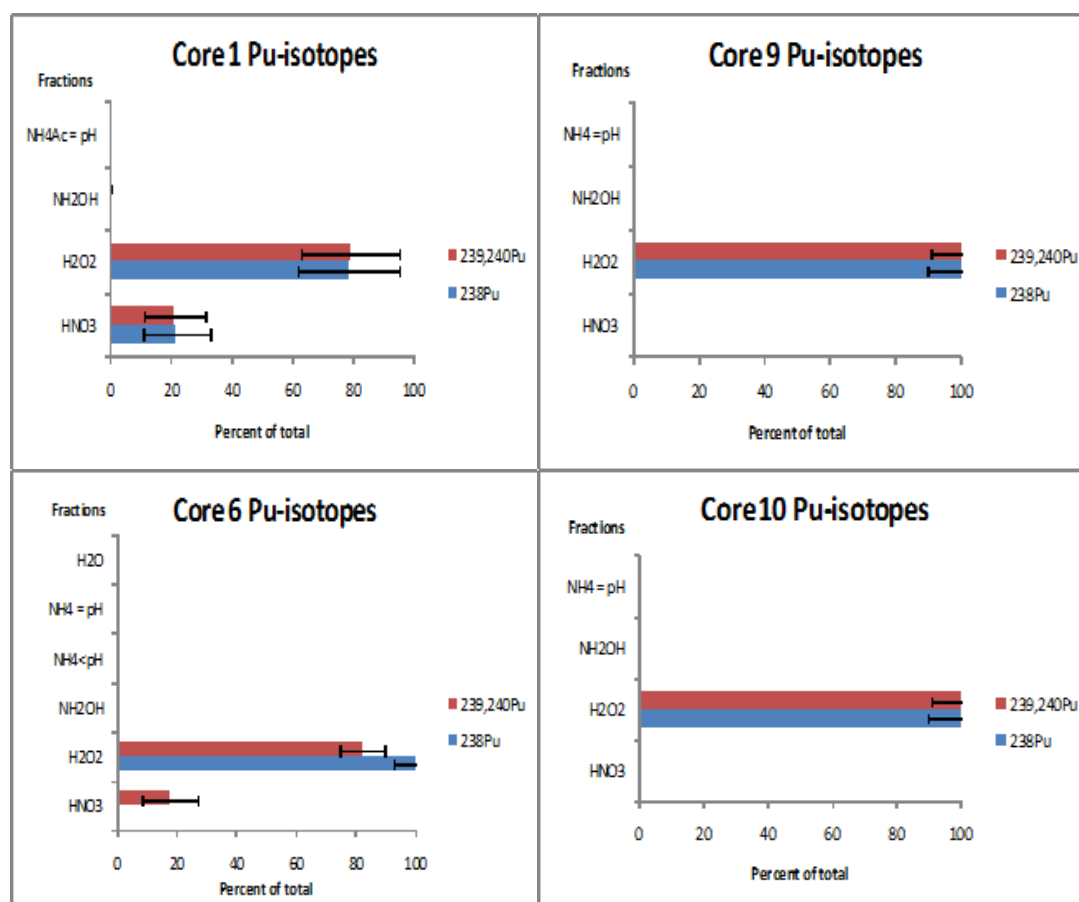


Figure 4. Sequential extraction results of Pu-isotopes in soil from outside (left, cores 1 and 6) and inside (right, cores 9 and 10) forest fire affected areas.

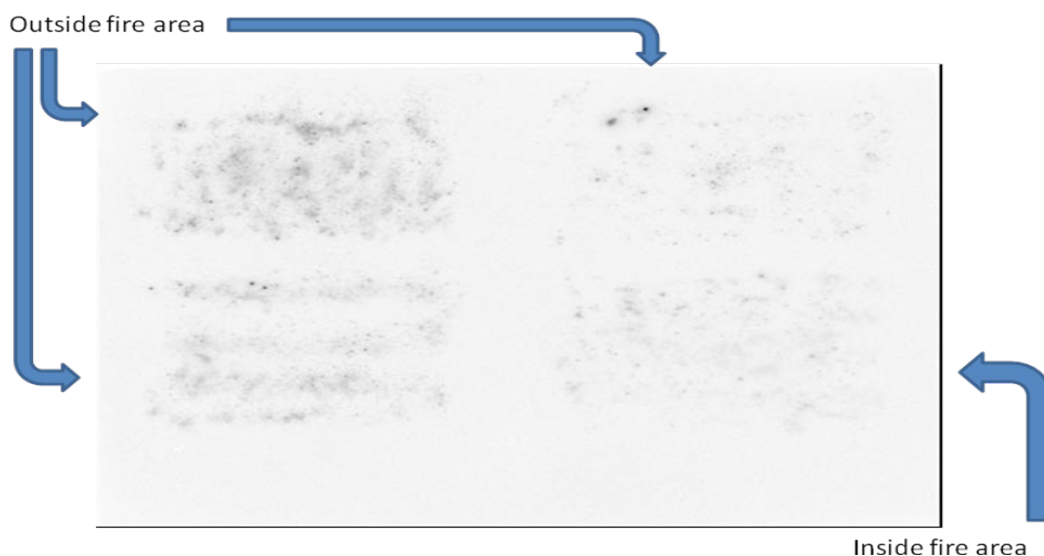


Figure 5. Digital autoradiography pictures of soils from different areas in the PSRER.

4. CONCLUSIONS

Even though no changes in speciation can be seen for Cs, a slight change for the Pu-isotopes can be seen comparing soils from inside and outside forest fire areas. Within the forest fire area, the Pu-isotopes are more bound to the more oxidisable form, and if incorporated in particles, this will be more susceptible to weathering after a fire.

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